

R E M A R K S

Applicants' present claim 3 is directed to a non-aqueous electrolyte for a lithium secondary battery to be used in combination with a positive electrode and a negative electrode capable of storing and releasing lithium, which comprises a non-aqueous solvent and a lithium salt dissolved therein,

wherein the non-aqueous solvent comprises:

- (a) at least one phosphate selected from the group consisting of
  - (a1) a chain state phosphate and (a2) a cyclic phosphate;
  - (b1) a cyclic carboxylate;
  - (c1) a vinylene carbonate compound and (c2) a vinylethylene carbonate compound.

The non-aqueous solvent recited in applicants' claim 3 thus contains (a) at least one phosphate selected from the group consisting of (a1) a chain state phosphate and (a2) a cyclic phosphate; and also contains (c1) a vinylene carbonate compound and (c2) a vinylethylene carbonate compound.

Claims 3, 4, 9 to 11, 13 to 15, 18 and 19 were rejected under 35 USC 103 as being unpatentable over Tan et al. (JP 11-

260401) in view of Yoshimura et al. (JP 4-087156) for the reasons set forth on pages 4 to 5 of the April 26, 2007 Office Action.

It was admitted in the April 26, 2007 Office Action that Tan et al. do not explicitly teach a vinylethylene carbonate compound.

Claims 20 to 27 were rejected under 35 USC 103 as being unpatentable over Tan et al. in view of Yoshimura et al. and further in view of Sonobe et al. (USP 5,527,643) and Kameda et al. (USP 6,632,569) for the reasons indicated on page 5, lines 4 to 11 of the April 26, 2007 Office Action.

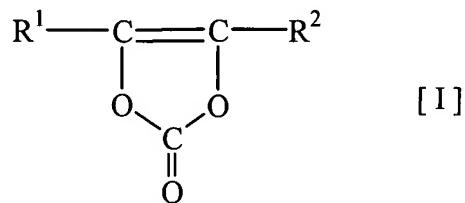
It was admitted in the July 12, 2006 Office Action that Tan et al. do not explicitly teach a graphite carbonaceous material having a first plane spacing  $d_{002}$  value, or a second carbonaceous material having a second plane spacing  $d_{002}$  value.

Claims 28 to 30 were rejected under 35 USC 103 as being unpatentable over Tan et al. in view of Sonobe et al. and Kameda et al. and further in view of Watanabe et al. (USP 6,682,856) for the reasons set forth in the last three paragraphs on page 5 of the April 26, 2007 Office Action.

It was admitted in the July 12, 2006 Office Action that Tan et al. do not explicitly teach the claimed surface area or particle diameter for the carbonaceous material or a metal selected from Sn, Si and Al.

Tan et al. (JP 11-260401 disclose the following:

[Claim 1] A non-aqueous electrolyte characterized by consisting of a non-aqueous solvent containing the vinylene carbonate derivative expressed by the following general formula [I], and a phosphoric ester compound, and an electrolyte:



In the above formula, even if R<sup>1</sup> and R<sup>2</sup> are mutually the same, they may differ from each other, and they include a hydrogen atom or an alkyl group having 1 to 3 carbon atoms.

In Tan et al., there is a description wherein the vinylene carbonate derivative represented by the formula [I] is added solely to an electrolyte containing a phosphoric acid ester compound.

Yoshimura et al. (JP 04-087156) disclose the following:

(1) A non-aqueous electrolyte battery which comprises a negative electrode comprising lithium or an alloy containing lithium, a positive electrode, and an electrolyte containing a solute and a solvent, and the above-mentioned solvent comprises at least one compound selected from the group consisting of a vinylethylene carbonate having an unsaturated carbon-carbon bond as a chain, 2-vinyl-1,3-dioxolane, 1,2-dimethoxyethylene, divinyl ether, N-vinylimidazole, vinyl amine and vinylcyclohexane.

Accordingly, Yoshimura et al. disclose to use a vinylethylene carbonate as a solvent.

The following is disclosed in claim 1 of Sonobe et al.

(USP 5,527,643:

"1. A carbonaceous electrode material for a non-aqueous solvent-type secondary battery, comprising a carbonaceous material having an average (002)-plane spacing  $d_{002}$  of 0.336-0.375 nm and a crystallite size in c-axis direction  $Lc_{(002)}$  of at most 50 nm, respectively, as measured by X-ray diffraction method, and an optically anisotropic texture showing a fine mosaic texture when observed through a polarizing microscope."

Accordingly, Sonobe et al. disclose only various physical properties of a specific negative electrode.

The following is disclosed in claim 1 of Kameda et al.

(USP No. 6,632,569):

"1. A carbonaceous material for electrode comprising a plane space  $d_{002}$  of a (002) plave less than 0.337 nm in an X-ray wide angle diffraction method, a crystallite size ( $Lc$ ) of 90 nm or higher, an R value, as a peak intensity ratio of a peak intensity ratio of a peak intensity of  $1360\text{ cm}^{-1}$  to a peak intensity of

1580 cm<sup>-1</sup> in a Raman spectrum in use of an argon ion laser, of 0.20 or higher, and a tap density of 0.75 g/cm<sup>3</sup> or higher."

Thus Kameda et al. disclose varioius physical properties of a specific negative electrode.

The following is disclosed in claim 1 of Watanabe et al. (USP 6,682,856) :

"1. A non-aqueous electrode secondary battery, comprising:  
a positive electrode, the positive electrode comprising a lithium-containing oxide;  
a negative electrode comprising a carbonaceous material and  
a non-aqueous electrolyte comprising a non-aqueous solvent, the  
non-aqueous solvent comprising at least one non-cyclic ester and  
an oxidation resistance improving agent, said oxidation  
resistance improving agent being an aryl-substituted alkyl  
compound in which 3 or more alkyl groups substituted with aryl  
groups."

Thus, Watanabe et al. disclose only an electrolyte containing a non-cyclic ester and an oxidation resistance improving agent (an aryl-substituted alkyl compound in which

three or more alkyl groups are substituted with aryl groups).

It is respectfully submitted that applicants' present claims are not obvious over Tan et al. (JP 11-260401) in view of other references cited by the Examiner as explained in detail below.

As described in the present specification, a non-aqueous electrolyte containing a phosphate as recited in applicants' present claims exhibits excellent flame retardancy (self-extinguishing property) and nonflammability (no flash point), so that it is possible to ensure safety of a lithium secondary battery. However, a phosphate is inferior in redox resistance to a conventionally used solvent and has a serious problem that it lowers the charge-discharge characteristics of a battery as described on page 3, lines 16 to 26 of the present specification.

In applicants' present claims, by using vinylene carbonate (hereinafter referred to as "VEC") and vinylethylene carbonate (hereinafter referred to as "VEC") in combination as components of the electrolyte, it was demonstrated in the DECLARATION UNDER 37 CFR 1.132 of Yasuyuki SHIGEMATSU dated October 5, 2006 and filed on October 16, 2006 that the problems involved in an electrolyte containing a phosphate could be solved. The above

fact deeply relates to a reductive decomposition potential of a phosphate and the film-forming potentials of VC and VEC as discussed below.

According to evaluations by the inventors after filing the present application, a reductive decomposition potential of trimethyl phosphate (hereinafter referred to as "TMP") can be estimated to be about 1.4V with a Li/Li<sup>+</sup> standard. On the other hand, VC forms a good film on a negative electrode, which film inhibits reductive decomposition at 0.8 to 1.3V (see the Journal of Electrochemical Society, 151, A1659-A1669 (2004) (a copy of which is of record), particularly see page A1660, right column, line 4 from the bottom to page A1662, left column, line 7 from the bottom). The above means that in an electrolyte containing TMP and VC, like in Tan et al. (JP 11-260401), charging proceeds to reach a potential of a negative electrode of 1.4V, and a reductive decomposition of TMP proceeds before formation of a film of VC. That is, in an electrolyte containing only TMP and VC and not containing VEC, an effect of VC is small, and no effect can be obtained in some cases. On the other hand, VEC has been known to have a film-forming property as in VC, and a film-

forming potential of VEC is nobler than that of VC. Moreover, a film-forming reaction of VEC starts from about 1.4V (or 1.35V) which is substantially the same as the reductive decomposition potential (see Electrochemistry Communications, 6, 126-131 (2004) (a copy of which is of record), particularly see page 127, right column, line 7 starting at "3. Results and discussion" to page 128, left column, line 4 from the bottom).

Accordingly, VEC and VC are clearly different from each other, and by carrying out a reductive decomposition of TMP simultaneously with a film formation of VEC, the reductive decomposition of TMP can be controlled. However, the film formed by VEC is not as good as that formed by VC, and when it is used as an additive of an electrolyte, its battery characteristics are not sufficient.

Thus, in applicants' present claims, by co-presenting VC in a non-aqueous electrolyte in combination with VEC and TMP, more practical battery characteristics can be obtained. That is, before TMP is decomposed by reduction, a reductive decomposition of TMP is stopped. Thus, a film formed only by VEC cannot be said to be good as that of VC. However, in the next step, a film

formation reaction by VC is expected. The film formed by VC has better qualities for a lithium secondary battery than the film formed by VEC, and improves battery characteristics. As a result, the battery characteristics of a battery containing both of VC and VEC are more improved than a battery containing either one of VC or VEC alone, and the former battery provides a practical use.

As can be clearly seen from the above explanation, when VC alone is used as in Tan et al. (JP 11-260401), desirable battery characteristics as afforded by applicants' present claims cannot be achieved. Moreover, in applicants' present claims, two kinds of solvents, both of which have a film-forming action, are not simply mixed in an electrolyte. When a phosphate is used as a solvent for an electrolyte, and yet it is contained in a higher concentration for the purpose of improving flame retardancy, battery characteristics can be synergistically heightened by co-presenting both of VC and VEC by the above-mentioned mechanisms. Accordingly, the effects of the electrolyte according to the applicants' present claims cannot be obtained only by combining an electrolyte of Tan et al. (JP 11-260401) using VC solely with

a publication which discloses an electrolyte containing a film-forming agent as a negative electrode.

Therefore, it is respectfully submitted that applicants' present claims cannot be arrived at by combining an electrolyte of Tan et al. (JP 11-260401) using VC solely with Yoshimura et al. (JP 4-0877156), which discloses simply a vinylethylene carbonate as a kind of a solvent.

Also, applicants respectfully submit that their present claims cannot be arrived at by combining an electrolyte of Tan et al. (JP 11-260401) using VC solely in combination with Yoshimura et al. (JP 4-0877156), and Sonobe et al. (USP 5,527,643) and Kameda et al. (USP No. 6,632,569), in the manner as set forth in the April 26, 2007 Office Action.

Moreover, applicants respectfully submit that applicants' present claims cannot be arrived at by combining an electrolyte of Tan et al. (JP 11-260401) using VC solely in combination with Yoshimura et al. (JP 4-0877156), and Sonobe et al. (USP 5,527,643) and Kameda et al. (USP 6,632,569), in view of Watanabe et al. (USP 6,682,856), in the manner as set forth in the April 26, 2007 Office Action.

Moreover, it is respectfully submitted that according to the mechanisms discussed above, it would be very surprising for one of ordinary skill in the art that battery characteristics (initial efficiency) are synergistically heightened 6% or more as seen in the aforesaid SHIGEMATSU DECLARATION. In particular, in Comparative example 1 which employed VC alone of Example 34 shown in Table 5 on page 4 of the aforesaid SHIGEMATSU DECLARATION, it was not charged nor discharged as shown in Table 6 on page 5 of the aforesaid SHIGEMATSU DECLARATION, but due to the co-presence of VC and VEC (i.e., Example 34), it showed a high charge-discharge efficiency. For example, Electrochimica Acta 52, (2007), 6006-6011 (a copy of which is of record) shows that by washing, heat-treating and coating graphite (a negative electrode active substance), an initial charge-discharge efficiency was improved 6 to 12%, as compared with that which had not been treated as shown in Table 1 on page 6008 therein. Such an increment of the initial charge-discharge efficiency is described as having been "greatly improved" at the column "4. Conclusions" on page 6010, right column thereof.

It is not taught or suggested by the cited references that the unexpected results of applicants' present claims can be obtained by combining an electrolyte disclosed in Tan et al. (JP 11-260401) using VC solely in combination with the other references cited by the Examiner.

Withdrawal of each of the prior art rejections as set forth in the April 26, 2007 Office Action is thus respectfully requested.

Reconsideration is requested. Allowance is solicited.

If the Examiner has any comments, questions, objections or recommendations, the Examienr is invited to telephone the undersigned at the telephone number given below for prompt action.

Respectfully submitted,



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